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ABSTRACT

Neodymium-doped fluorophosphate glass is a laser material newly-developed for use in high power laser fusion systems. The low refractive index ($n_d \sim 1.45$) and low dispersion (Abbe number $\sim$90) of fluorophosphate glasses give them the properties of low nonlinear refractive indices and long Nd^{3+} fluorescence lifetimes, which are desirable for the high power laser applications. We have measured the intensity gain of 1.052 and 1.064 nm laser light produced by flashlamp-pumped fluorophosphate glass amplifiers, varying in size from 4 cm to 34-cm clear aperture. The measured gains are compared to those measured in other laser glass types and to those predicted from the spectroscopic properties of Nd^{3+}. We estimate that the peak cross section for the $^4F_{3/2} \rightarrow {}^4I_{11/12}$ transition in commercial fluorophosphate laser glasses is $\sim 2.2 \times 10^{-20} cm^2$.

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I. Introduction

Research into the possibility of using inertial confinement fusion as an energy source requires high-power high-energy lasers.[1] The workhorse of this research effort is the Nd-glass laser. The Shiva laser at Lawrence Livermore Laboratory, operational since November 1977, can provide on a target 30 TW of 1064 nm light in a pulse < 1 ns wide or 15 kJ for pulse widths > 1 ns. This laser uses Nd^{3+}-doped silicate glass. Under construction is the Nova laser, designed to be ten times more powerful than Shiva. To meet the higher power requirements of Nova, a new laser glass, based on fluorophosphate compositions has been developed. We present the optical and mechanical properties of this new glass and the results of small signal gain measurements in laser amplifiers. Some properties of Nd-doped fluorophosphate glasses were previously reported by Deutschbein et al.[2]

Fluorophosphate glass is needed for high power lasers because it has a low nonlinear refractive index, n_2. The nonlinear index, which is due to the dependence of the refractive index on intensity, results in the growth of spatial inhomogeneities in laser beam profile.[3] These spatial variations cause loss of energy that can be focussed on a fusion target. Quantitatively, a factor of two more focusable power can be propagated though a fluorophosphate laser amplifier chain than through an equivalent silicate chain. We illustrate this problem in Figure 1, where we compare calculations of a spatial beam profile after propagation through silicate and fluorophosphate glass at two different power levels. Fluorophosphate glass has a n_2 of $\sim 0.55 \times 10^{-13}$ esu, whereas the n_2 of silicate glass is $\sim 1.5 \times 10^{-13}$ esu, more than a factor of 2.5 larger. At low powers any spatial intensity variations in the beam, due to obscurations (dirt, damage, etc.), grow slowly as the pulse propagates through the glass. However, at high powers the growth of beam spatial modulations is much slower in fluorophosphate glasses with their lower n_2 than in silicates.

II. Properties of Fluorophosphate Glass

The nonlinear indices of fluorophosphate glasses are lower than those of silicates or phosphates used in present laser fusion systems because the hyperpolarizability of the fluorine anion is smaller than that of oxygen.[4] A typical fluorophosphate glass composition listed in Table I shows that it is mixed fluoride with very little P_2O_5 glass former. Even lower n_2's can be obtained in pure fluoride glasses, such as the BeF_2-based glasses;[5] however, their toxicity and lack of development make them undesirable for use in the Nova laser.

The optical and physical properties of fluorophosphate glasses are compared to those of typical phosphate and silicate laser glasses in Table II. The low n_2 and small stress-optic coefficient give fluorophosphates a significant advantage over the other glass types. On the other hand, its high thermal expansion coefficient and softness means that more care must be taken in handling and finishing it.

Our primary interest is in the spectroscopic properties of Nd^{3+} in fluorophosphate glasses because they relate to laser performance. From the measured absorption spectra, fluorescence spectra, fluorescence decay, and Nd concentration we obtained the spectroscopic values listed in Table III. We calculated the emission cross sections and radiative lifetimes using a Judd-Ofelt analysis of transition probabilities in rare-earth ions.[6] These values are considered to be $\pm 10\%$ accurate and depend directly on the accuracy of the measured Nd concentration, which is determined chemically.[7] The fluorescence decay is nonexponential in form because of the inhomogeneous nature of the Nd^{3+} sites in the glass, each ion being in a different local environment. We define the decay time as the first e-folding time. The longer lifetime of fluorophosphate glasses is an advantage in amplifiers in which the flashlamp pump pulse is $\sim 600\ \mu s$. The effective linewidth is the integral over wavelength of the fluorescence intensity, normalized to the peak intensity.

The mixed anion nature of fluorophosphate glass is reflected in the fluorescence spectrum of this glass which spans the wavelength region between the 1054 nm fluorescence peak of phosphate glasses and the 1047 nm peak of BeF_2-based pure fluoride glasses.[8] The effective linewidth, which partially characterizes the inhomogeneity of the Nd^{3+} spectra, is larger for fluorophosphate glass than for phosphate ($\sim$26 nm) or BeF_2-based ($\sim$23 nm) glasses.

III. Gain Measurements

In a homogeneous laser material the small signal gain coefficient (α) is equal to the emission cross section (σ) times the number of excited Nd^{3+} ions (N^*) i.e.,

$$\alpha = \sigma N^* \tag{1}$$

However, each Nd ion in glass is in a different local environment, which results in a distribution of cross section due to variations in the line strength, peak wavelength, and homogeneous linewidth of the individual transitions. Thus,

$$\alpha = \sum_i \sigma_i n_i^* \simeq \bar{\sigma} N^* \tag{2}$$

where the summation is over the individual Nd sites and $\bar{\sigma}$ is the average cross section. Thus, the inhomogenous nature of glass is not observable in the small signal gain. However, under gain saturation conditions produced by high energy input pulses, the distribution of cross sections become important.

In our measurements amplifier gains are determined at very low power levels ($\leq$ 1 watt). The relative cross sections of different glasses can be derived from measurements of their gain in amplifiers using an amplifier and flashlamp pumping model to obtain the relative N^*'s.

The gain of fluorophosphate glass in disk amplifiers was measured at two different apertures, 4.8 cm and 34 cm. The small disks have dimensions 4.8 x 8.4 x 1.5 cm and are surrounded on their periphery by an index matched, 1050 nm-absorbing solution of $ZnCl_2$, ZnI_2 or $NiCl_2$. (A liquid or solid material that absorbs at the lasing wavelength and is index matched to the laser glass is known as an edge cladding. Edge claddings on laser disks are necessary to prevent stored energy loss due to laser oscillations in the plane of disk or to amplified spontaneous emission.)[9] The disks are placed in an amplifier containing the liquid edge cladding holders and flashlamps. The large amplifier consists of two solid edge clad 34 x 66 x 5 cm disks surrounded by flashlamps and silvered refelctors. The experimental apparatus and measurement techniques are described in Reference 9. The experimental procedure eliminates the effects of passive loss in the amplifier and thus, measures the gross gain characterized by Equation 1, not net gain. We used a Nd: YAG cw laser operating at 1052 nm or 1064 nm to measure the gain near the peak spectral peak of fluorescence. We plot the experimental values in Figure 2. Each point is the averaged value of individual measurements with the error bars indicating the standard deviation.

We compare also in Figure 2 the measured data with the gain coefficient predicted from a computer code that models laser amplifier performance.[9] Because of the time and expense involved in testing different types of laser glass, we employ a computer model to predict amplifier performance. This amplifier model uses as input:

1) an empirical time dependent model for conversion of electrical energy to flashlamp light.
2) the measured absorption of the glass
3) the fluorescence decay in the glass
4) a model for the amplified spontaneous emission loss
5) the calculated emission cross section
6) the cavity efficiency

(A detailed description of the above model and its application to amplifier performance is given in 9.) The cavity efficiency is an empirical parameter that we set by measuring the gain of a reference glass, ED-2 silicate laser glass. With the above parameters, we can predict the relative performance of a glass.

The measured fluorophosphate gains fall $\sim$10% lower than predicted on the basis of spectroscopic measurements. Because the most uncertain parameters in our calculations are the cross sections, we suspect that the relative calculated cross sections of fluorophosphate and ED-2 are not correct. Our suspicion is supported by the observation that the calculated radiative lifetime for the fluorophosphate glass is shorter than measured decay time at low Nd concentrations, for which the radiative quantum efficiency should be high. Because the radiative quantum efficiency must be < 1, one expects the measured decay time to be shorter than the radiative lifetime. If we lower the calculated strength of the ${}^4F_{3/2} \rightarrow {}^4I_J$ transitions by 15%, the radiative lifetime will be in agreement with the measured low Nd^{3+} concentration decay time, and the cross section will be lower by 15% ($\sigma \sim 2.2 \times 10^{-20}$ cm^2) in agreement with the gain measurements. Thus, our evidence indicates that the Judd-Ofelt analysis may give an overestimate for the cross section when applied to fluorophosphate glasses.

IV. Conclusions

Fluorophosphate glass is an excellent material to employ in high power laser systems because of its low nonlinear refractive index. We have estimated the peak emission cross section of the ${}^4F_{3/2} \rightarrow {}^4I_{11/12}$ transition in fluorophosphate glass to be 2.2×10^{-20} cm^2 using ED-2 silicate laser glass as a reference ($\sigma = 2.7 \times 10^{-20} cm^2$).

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We thank W. Warren for providing his results on nonlinear propagation of high power laser beams through glass. We also appreciate useful discussions with M. J. Weber.

TABLE I

TYPICAL FLUOROPHOSPHATE LASER GLASS COMPOSITION
(in Mole %)

$Al(PO_3)_3$	5
AlF_3	32
NaF	12
MgF_2	8
CaF_2	27
SrF_2	8
BaF_2	8

TABLE II

OPTICAL AND PHYSICAL PROPERTIES OF COMMERCIAL LASER GLASSES

Glass Glass Type	LG-812[a] Fluorophosphate	LHG-8[b] Phosphate	ED-2[c] Silicate
Refractive index at 589 nm, n_D	1.431	1.531	1.572
Abbe number, ν	93.1	70.2	57.5
Nonlinear refractived index (10^{-13} esu)	0.55	1.1	1.5
Index temperature coefficient, $\delta n/\delta T$ 10^{-6} $^{o}C^{-1}$ @ 633 nm	-7.5	-5.3	2.9
Optical Path Change with temperature, $\frac{\delta S}{\delta T}$ (10^{-6} $^{o}C^{-1}$) @ 633 nm	1.4	0.6	8.0
Stress-optic coefficent (nm $\cdot$ cm $\cdot$ kg^{-1}) at 633 nm	0.91	1.93	2.06
Expansion coefficient (10^{-6} $^{o}C^{-1}$)	14.6	11.2	9.3
Transformation temp (^{o}C)	407	485	468
Density (g /cm^3)	3.19	2.85	2.54
Knoop hardness (kg/mm^2) @ 100g	330	321	599
Young's modulus (kg mm^{-2})	7700	5109	9189
Shear modulus (kg mm^{-2})	2890	2030	3698

a Schott Optical Glass Inc; equivalent glasses are manufactured by Owens Illinois (E-309) and Hoya (LHG-10).
b Hoya Corporation; equivalent glasses are manufactured by Schott (LG-700), Kigre (Q-88) and Owens Illinois (EV-4).
c Owens-Illinois; an equivalent glass is LSG-91H (Hoya).
d Estimated

TABLE III

SPECTROSCOPIC PROPERTIES OF THE ND^{3+} $^4F_{3/2} \rightarrow {}^4I_{11/2}$ TRANSITION IN COMMERCIAL LASER GLASSES

Glass Glass Type	LG-812[a] Fluorophosphate	LHG-8[b] Phosphate	ED-2[c] Silicate
Emission cross section[d] ($10^{-20}cm^2$)	2.6	4.0	2.7
Radiative lifetime[d] (μ s)	495	338	359
Decay time for low low Nd^{3+} concentrations (μ s)	550	380	360
Peak Fluorescence Wavelength (nm)	1051	1053	1061
Full width at half maximum (nm)	26.1	21.8	27.8
Effective linewidth (nm)	31.0	25.9	34.4

a Schott Optical Glass, Inc.
b Hoya Corporation
c Owens Illinois, Inc.
d Calculated from a Judd-Ofelt analysis

FIGURE CAPTIONS

FIGURE 1 Intensity profile of a laser beam before and after propagation through 14 cm of silicate or fluorophosphate glass at two different intensity levels. These profiles were generated by a computer code that models light propagation in nonlinear media.

FIGURE 2 Measured small signal gain coefficient of a) two 34 x 66 x 5 cm disks E-309 fluorophosphate laser glass at 1052 nm and b) two 4.8 x 8.4 x 2.0 cm disks of LHG-10 fluorophosphate glass at 1064 nm as a function of capacitor bank energy. The predicated curves are obtained from the measured spectroscopic data and a computerized model of the amplifers.

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FIGURE 1

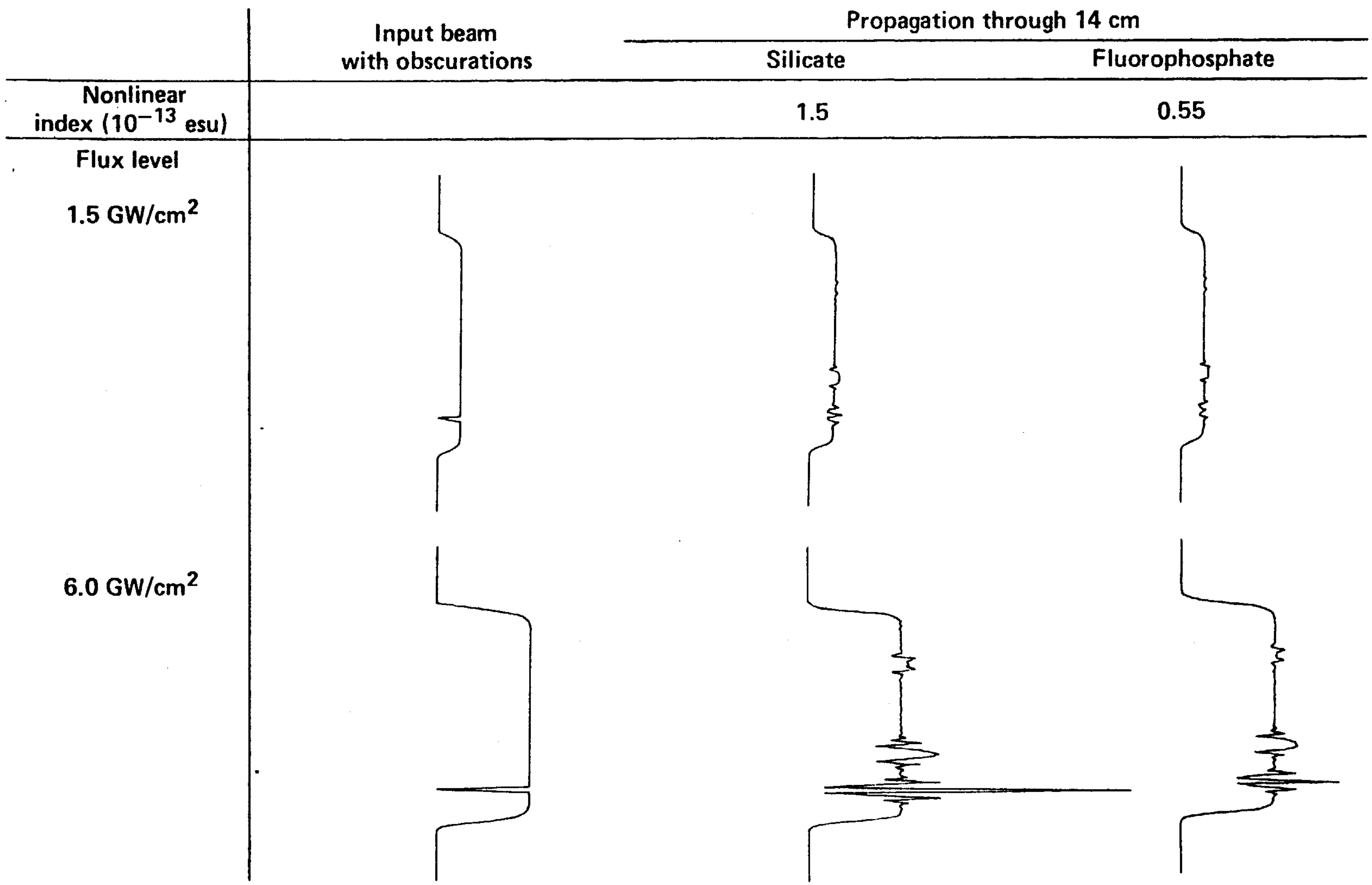
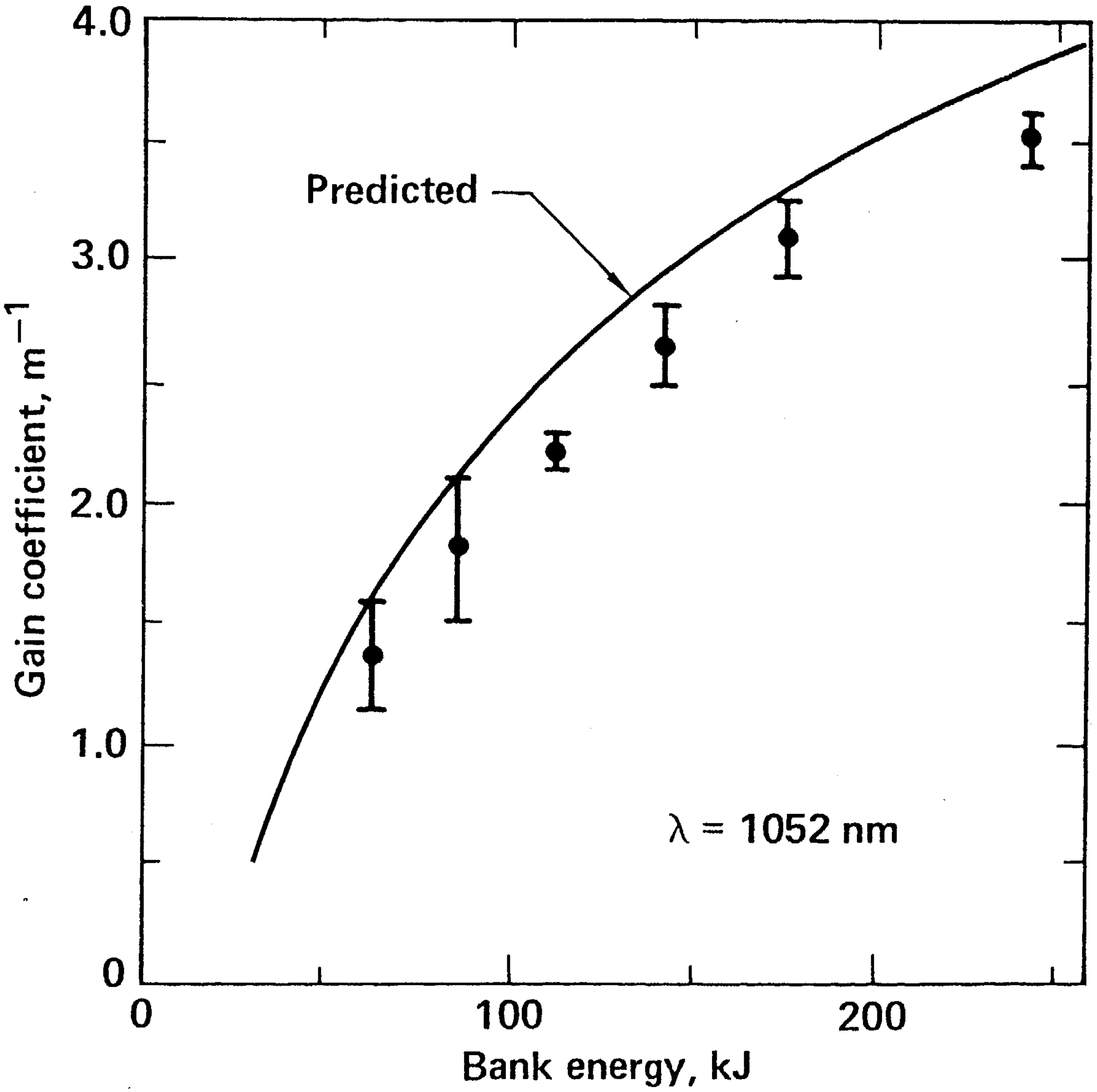
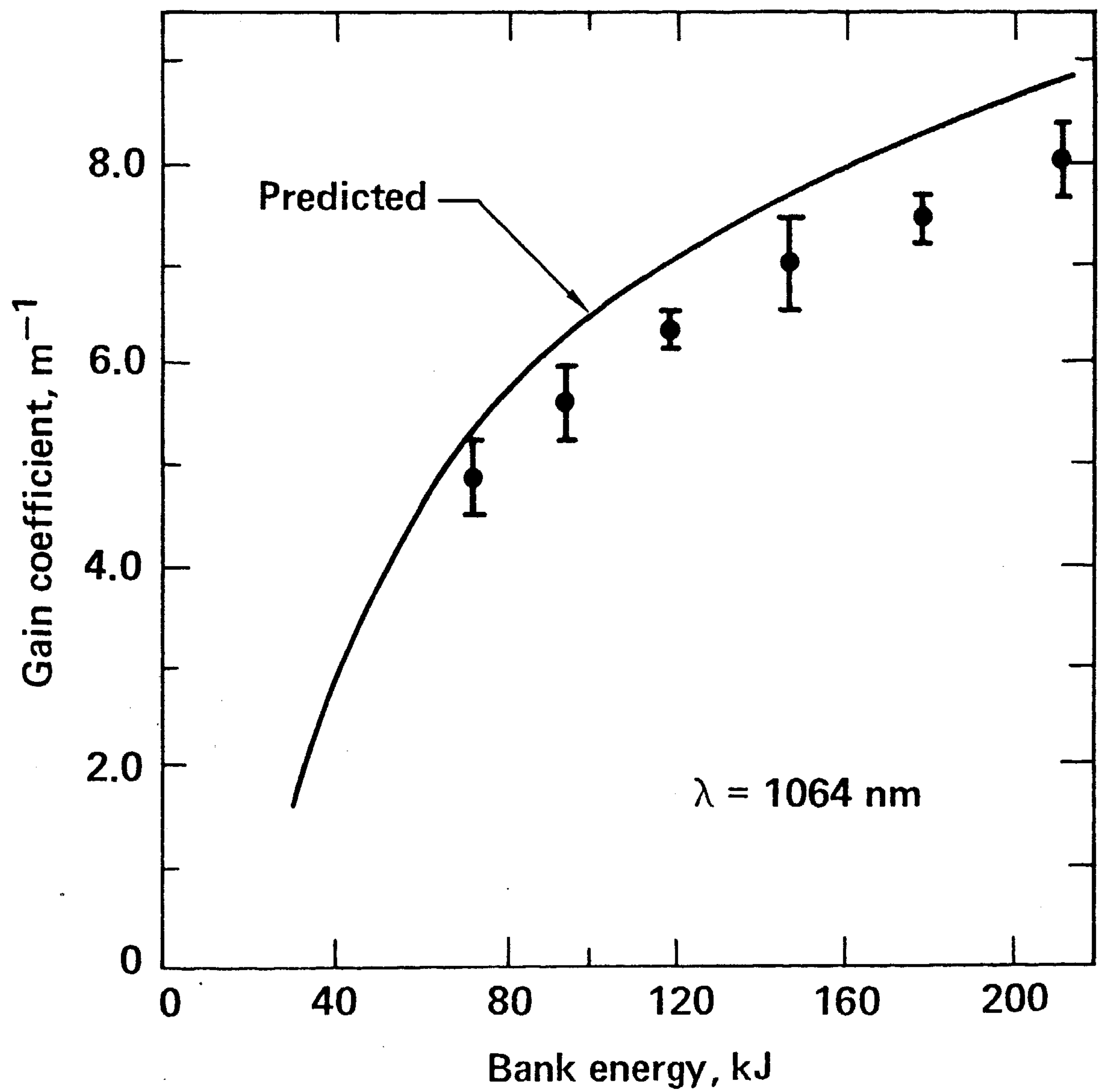

Input beam
with obscurations
Propagation through 14 cm
Silicate
Fluorophosphate
Nonlinear
index (10−13 esu)
1.5
0.55
Flux level
1.5 GW/cm2
6.0 GW/cm2

FIGURE 2a



4.0
3.0
2.0
1.0
0
Gain coefficient, m^{-1}
Predicted
λ = 1052 nm
0
100
200
Bank energy, kJ

<u>FIGURE 2b</u>


Predicted
λ = 1064 nm
Gain coefficient, m⁻¹
8.0
6.0
4.0
2.0
0
0
40
80
120
160
200
Bank energy, kJ